

# Angle-Resolved Photoemission Spectroscopy of the Antiferromagnetic Superconductor $\text{Nd}_{1.87}\text{Ce}_{0.13}\text{CuO}_4$ : Anisotropic Spin-Correlation Gap, Pseudogap, and the Induced Quasiparticle Mass Enhancement

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We performed high-resolution angle-resolved photoemission spectroscopy on  $\text{Nd}_{1.87}\text{Ce}_{0.13}\text{CuO}_4$ , which is located at the boundary of the antiferromagnetic (AF) and the superconducting phase. We observed that the quasiparticle (QP) effective mass around  $(\pi, 0)$  is strongly enhanced due to the opening of the AF gap. The QP mass and the AF gap are found to be anisotropic, with the largest value near the intersecting point of the Fermi surface and the AF zone boundary. In addition, we observed that the QP peak disappears around the Néel temperature ( $T_N$ ) while the AF pseudogap is gradually filled up at much higher temperatures, possibly due to the short-range AF correlation.

Since the discovery of cuprate high-temperature superconductors (HTSCs), intensive experimental and theoretical studies have been performed to elucidate the origin and mechanism of the anomalously high superconducting (SC) transition temperature. It is now widely accepted that electrons or holes doped into the parent Mott insulator interact antiferromagnetically with each other on the quasi-two dimensional  $\text{CuO}_2$  plane. Although it has been suggested that the antiferromagnetic (AF) interaction plays an essential role for pairing of electrons (holes) in the SC state, it is still unclear how the antiferromagnetism interplays with the superconductivity at the microscopic level. This problem is a central issue not only in HTSCs but also in other exotic superconductors such as heavy-fermion and organic-salt superconductors. In the phase diagram of electron-doped cuprates, the AF and SC phases are adjacent to each other or somewhat overlap at the boundary, in contrast to the hole-doped case where the two phases are well separated [1]. The proximity or overlapping between the AF and SC phases in the electron-doped cuprates yields a good opportunity for studying the interplay between the AF interaction and the superconductivity [2, 3, 4, 5]. In fact, a recent elastic neutron-scattering experiment reported a competitive nature between the AF long-range order and the superconductivity [2]. On the other hand, an inelastic neutron scattering experiment observed coexistence of the gapped commensurate spin fluctuation and the superconductivity [3]. In contrast to these intensive studies on the “ $q$ -resolved” spin dynamics by neutron scattering, a limited number of photoemission studies on the “ $k$ -resolved” electronic structure have been reported for electron-doped cuprates [6, 7]. The  $k$ -dependence of the AF correlation effect on the electronic structure is essential to understand the interplay between the antiferromagnetism and the superconductivity.

In this Letter, we report high-resolution angle-resolved photoemission spectroscopy (ARPES) on electron-doped

cuprate  $\text{Nd}_{1.87}\text{Ce}_{0.13}\text{CuO}_4$  (NCCO,  $x = 0.13$ ) located at the phase boundary between the AF and SC phases. We found the mass-renormalized quasiparticle (QP) state near  $(\pi, 0)$ , which gradually evolves into the high-energy gap [6] around the hot spot. The observed continuous evolution of the electronic structure near the Fermi level ( $E_F$ ) as a function of momentum ( $k$ ) is explained basically in terms of the band folding caused by the AF ordering. However, we also found a deviation from this simple picture in the  $k$ -dependence of the AF-gap size indicative of non-uniform AF scattering in  $k$ -space. We observed that the QP spectrum shows remarkable temperature dependence in accordance with the spin correlation.

High-quality single crystals of NCCO ( $x = 0.13$ ) were grown by the traveling solvent floating zone method and were heat-treated in Ar-gas flow at 900°C for 10h. The  $\mu\text{SR}$  and magnetic susceptibility measurements show that the crystal is antiferromagnetic below 110 K ( $T_N$ , Néel temperature) and shows a signature of superconductivity below 20 K ( $T_c$ ) [8]. ARPES measurements were performed with GAMMADATA-SCIENIA SES200 and SES2002 spectrometers at Tohoku University and the undulator 4m-NIM beamline of Synchrotron Radiation Center, Wisconsin, respectively. We used monochromatized He I $\alpha$  resonance line (21.218 eV) and 22-eV photons to excite photoelectrons. The energy and angular (momentum) resolutions were set at 11 meV and 0.2° (0.01Å<sup>-1</sup>), respectively. A clean surface of sample for ARPES measurements was obtained by *in-situ* cleaving along the (001) plane. The Fermi level of sample was referred to that of a gold film evaporated onto the sample substrate.

Figure 1 shows the plot of ARPES intensity near  $E_F$  at 30 K as a function of the two-dimensional wave vector to illustrate the Fermi surface (FS) of NCCO ( $x = 0.13$ ). Bright areas correspond to the experimental FS. We normalized the intensity with respect to the highest

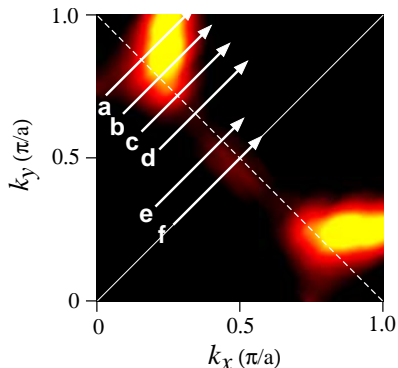


FIG. 1: Plot of near- $E_F$  ARPES intensity at 30 K integrated within 25 meV with respect to  $E_F$  and symmetrized with respect to the  $(0, 0)$ - $(\pi, \pi)$  nodal line. Arrows denoted by  $a$ - $f$  show cuts where detailed ARPES measurements shown in Fig. 2 were done.

binding energy of spectrum (400 meV) [9]. We find in Fig. 1 that the ARPES intensity at  $E_F$  shows a characteristic  $k$ -dependence while the experimental FS looks circle-like centered at  $(\pi, \pi)$  as predicted from the LDA band calculation [10]. On the experimental FS, the strongest ARPES intensity appears near  $(\pi, 0)$ , and a weak but observable intensity is seen around  $(\pi/2, \pi/2)$ , while there is negligible or no intensity between these two momentum regions. It is noted that the area with negligible ARPES intensity on the FS coincides with the hot spot, namely the intersecting point of the LDA-like FS and the AF zone boundary. A similar ARPES-intensity modulation has been reported in the case of  $x = 0.15$  with different photon energies [6], suggesting that the observed intensity modulation is not due to the matrix-element effect.

Figure 2 shows the ARPES spectra near  $E_F$  measured along several cuts across the FS (cuts  $a$ - $f$  in Fig. 1) and the corresponding band dispersions derived from the spectra. Near the  $(\pi, 0)$  point (cuts  $a$ - $c$ ), we find two separated band dispersions; one is a very steep band dispersion located below  $\sim 0.1$  eV and another is a flat band very close to  $E_F$ . The strong ARPES intensity on the FS near the  $(\pi, 0)$  point as shown in Fig. 1 is due to this flat band located very close to  $E_F$ . As seen in Fig. 2, the presence of two separated bands in the same momentum region produces the characteristic “peak-dip-hump (PDH)” structure in the ARPES spectrum measured near the Fermi vector ( $k_F$ ). The band near  $E_F$  becomes flatter and the intensity is weakened on going from cut  $a$  to cut  $c$ , namely on approaching the hot spot. At the same time, the energy separation between the peak and the hump gradually increases. In cut  $d$ , which passes the hot spot, the peak near  $E_F$  almost disappears and as a result an energy gap of about 100 meV opens between  $E_F$  and the lower-lying steep band. The energy gap becomes gradually small on approaching the nodal line and finally the steep band appears to al-

most touch  $E_F$  in cut  $f$ , as evidenced by the recovery of the Fermi-edge-like structure in the spectrum. Here, it is noted that the change of band dispersions among different cuts is continuous, indicating that the PDH structure in the ARPES spectra in cuts  $a$ - $c$  has a same origin as the energy gap in cuts  $d$  and  $e$ .

The opening of a large energy gap at  $E_F$  in cut  $d$  is attributed to the AF spin-correlation, since it is located at the intersecting point of the “original” FS and the “shadow” FS produced by the AF interaction [6]. The present ARPES results in Fig. 2 clearly show that the gap at the hot spot is smoothly connected to the two separated bands near the  $(\pi, 0)$  point, suggesting the effect of the AF correlation to modify the band dispersion. We show in Fig. 3 a schematic diagram to explain how the QP dispersion is modified by the AF electron correlation. It is reminded that the intersecting point between the original band and the shadow band folded back into the magnetic Brillouin zone is always on the diagonal line  $[(\pi, 0)$ - $(0, \pi)]$ , and more importantly, the intersecting point is *below*  $E_F$  at  $(\pi, 0)$  and *above*  $E_F$  at  $(\pi/2, \pi/2)$  in the presence of a nearly half-filled circle-like FS centered at  $(\pi, \pi)$  as shown in Fig. 3. The relative position of this intersecting point with respect to  $E_F$  plays an essential role in characterizing the band dispersion and the ARPES intensity on the FS. In the case I in Fig. 3, where the intersecting point is below  $E_F$ , the strong AF scattering splits the original dispersion into two pieces above and below the intersecting point, respectively, producing an energy gap between the two separated bands. It is expected that the occupied band just below  $E_F$  is strongly bent and the QP effective mass is remarkably enhanced in this momentum region. This effect becomes stronger when one approaches the hot spot, because the intersecting point with the strongest AF scattering is gradually shifted to  $E_F$ . In the case II, where the intersecting point is just on  $E_F$ , the AF scattering eliminates the electronic states at  $E_F$ , producing a large energy gap at  $E_F$ . Finally in the case III, where the intersecting point is above  $E_F$ , the AF interaction affects the band dispersion mainly in the unoccupied states, leaving the original band dispersion in the occupied states almost unaffected. We find that the gross feature of band dispersions in different cuts in Fig. 2 shows a good agreement with this simple picture. For example, cuts  $a$ ,  $d$ , and  $f$  correspond to the cases I, II, and III, respectively. Thus the observed heavy-mass QP state around  $(\pi, 0)$  and the  $k$ -dependence of QP dispersion are well explained in terms of the effect from the AF correlation. The continuous evolution of band dispersion along the FS shown in Fig. 2 strongly suggests the AF origin of the energy gap and the resulting mass-enhancement in NCCO.

We find in Fig. 2 that the energy separation between the peak and the hump (namely the energy separation between the upper and the lower bands separated by the AF correlation) gradually increases from cut  $a$  ( $\sim 50$  meV)

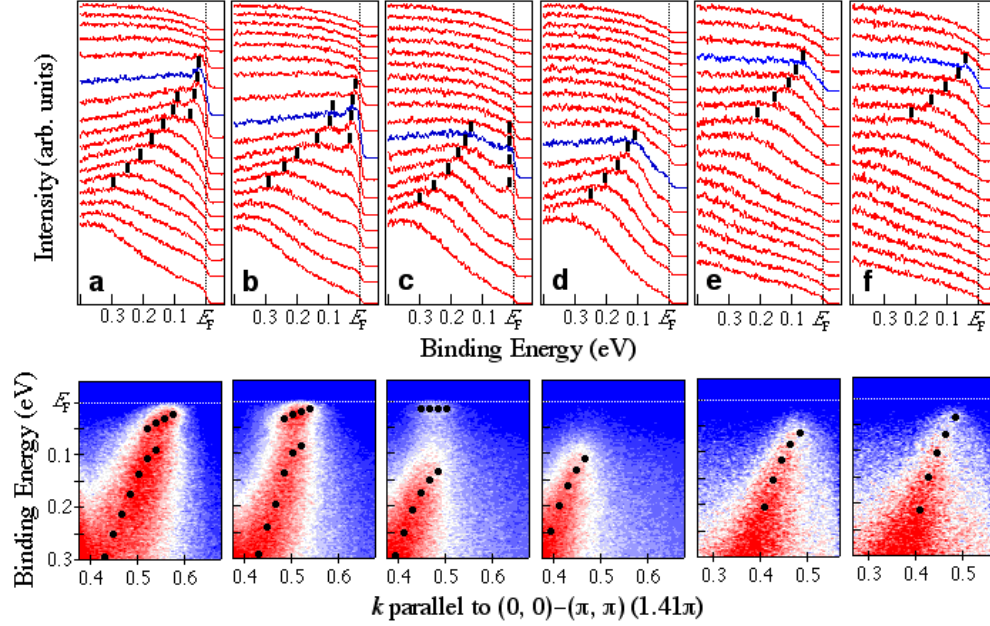


FIG. 2: Upper panel: ARPES spectra of NCCO ( $x = 0.13$ ) measured at 30 K along several cuts parallel to the  $(0, 0)$ - $(\pi, \pi)$  direction shown by arrows in Fig. 1. Blue spectra are at the Fermi surface. Lower panel: ARPES-intensity plot as a function of the wave vector and binding energy, showing the experimental band dispersion. Peak positions in ARPES spectra are shown by bars and dots.

to cut  $c$  ( $\sim 120$  meV). This  $k$ -dependence of the AF gap is not necessarily obvious in the simple picture in Fig. 3 and may suggest that the strength of the AF scattering has momentum dependence, with the stronger amplitude close to the hot spot. A recent tunneling spectroscopy reported a pseudogap comparable in the size to the superconducting gap, suggesting the second order parameter hidden within the superconducting state in electron-doped HTSCs [11]. However, the one-order smaller energy scale compared to the AF gap suggests the different nature between these two gaps.

The mass-enhancement effect and the PDH structure in Fig. 2 look similar to those in hole-doped HTSCs, which have been interpreted with some corrective modes such as the magnetic-resonance mode [12]. However, such arguments is not applicable to the electron-doped case, because the QP state is clearly observed even above  $T_c$ . As described above, the mass-enhancement in NCCO is due to the band folding caused by the AF order/fluctuation. In this case, the energy separation between the *peak* and the *dip* does not reflect the energy of collective mode, but the separation between the *peak* and the *hump* is related to the AF exchange interaction. It is also remarked here that the  $k$ -region where the heavy-mass QP state is observed coincides with the  $k$ -region where a large  $d$ -wave superconducting gap opens [13]. This suggests that the superconductivity in electron-doped HTSCs occurs in the antiferromagnetically correlated QP state [14]. The present experimental

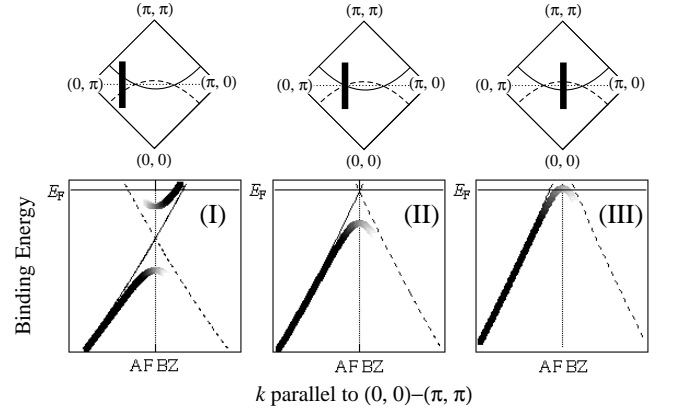


FIG. 3: Schematic diagram to explain how the QP dispersion is modified by the AF correlation for three different cases. In case I, the original QP band (thin solid line) and the shadow band (thin broken line) intersect each other below  $E_F$ . In cases II and III, the intersecting point is at and above  $E_F$ , respectively. Thick solid lines show the QP dispersions modified by the AF correlation.

result that the QP effective mass at  $E_F$  and the AF gap increase as moving away from  $(\pi, 0)$  suggests a slight deviation in the SC order parameter from the simple  $d_{x^2-y^2}$  symmetry,  $\Delta(k) \propto \cos(k_x a) - \cos(k_y a)$ , in NCCO [15, 16].

Next, we discuss how the heavy-mass QP state at cuts  $a$ - $c$  in Fig. 2 changes as a function of temperature. Figure 4 shows the temperature dependence of ARPES spectrum measured at a point on the FS where the PDH

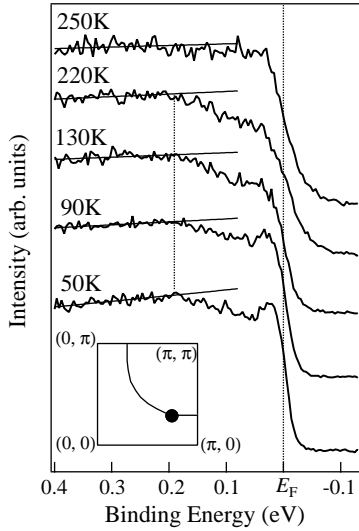


FIG. 4: Temperature dependence of ARPES spectrum of NCCO ( $x = 0.13$ ) measured at a point on the Fermi surface shown by a filled circle in the inset, where the PDH structure is clearly observed. Solid straight lines on the spectra show the linear fits to the high-energy region (0.2 - 0.5 eV).

structure is observed (see the inset to Fig. 4). At low temperatures (50 and 90 K) below  $T_N$  (110K), we clearly find a relatively sharp QP peak at  $E_F$  and a broad hump at 190 meV in the ARPES spectra, which are ascribed to the upper and lower pieces of the renormalized QP band, respectively. On increasing the temperature across  $T_N$ , the QP peak at  $E_F$  becomes substantially broadened and almost disappears at 130K. However, the suppression of the spectral weight near  $E_F$  ( $E_F - 0.2$  eV), which defines “a large pseudogap”, is still seen in the spectra at 130 K and 220 K. This suggests that while the long-range spin ordering disappears at  $T_N$ , the short-range spin-correlation survives even above  $T_N$ , affecting the electronic structure near  $E_F$ . It is remarked that the energy of hump (190 meV) does not change with temperature. On further increasing the temperature, the pseudogap is totally filled-in in the spectrum at 250 K, suggesting that the short range AF correlation disappears at around this temperature. The optical conductivity experiment has reported that a pseudogap-like suppression starts to develop in the energy-range lower than 0.18 eV at 190 K for  $x = 0.125$  [17], consistent with the present study. Further, the optical conductivity and the Raman experiments have reported the simultaneous evolution of both the low-energy Drude-like response and the high-energy gap on decreasing the temperature [17, 18]. This shows a good correspondence to the gradual development of the QP peak at  $E_F$  in ARPES spectrum at low temperatures. The reported sharpening of the low energy optical response in NCCO [17, 18] is well explained in terms of the QP mass-enhancement due to the AF correlation.

In conclusion, we have performed high-resolution ARPES study on electron-doped cuprate  $\text{Nd}_{1.87}\text{Ce}_{0.13}\text{CuO}_4$ . We observed a systematic variation of the QP band dispersion as a function of momentum in addition to the characteristic ARPES-intensity modulation on the FS. These experimental results are well explained in terms of the  $k$ -dependent band-folding effect due to the AF ordering. We observed that the effective QP mass around  $(\pi, 0)$  is strongly renormalized due to the opening of the AF gap. The QP mass and the AF gap are anisotropic, with the largest value near the hot spot, and with a smaller value in the vicinity of  $(\pi, 0)$ , suggesting the stronger AF scattering at the hot spot than at  $(\pi, 0)$ . Temperature-dependent measurements show that the QP peak gradually weakens on increasing temperature and disappears at around the Néel temperature, while the AF pseudogap defined by the hump structure is seen at temperatures much higher than  $T_N$ , suggesting that the short-range AF correlation survives well above  $T_N$ .

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